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NOTIFICATION OF ELECTION

(PCT Rule 61.2)

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| Date of mailing (day/month/year) 20 June 2000 (20.06.00) | in its capacity as elected Office | | | | |
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| International application No. PCT/US99/24533 | Applicant's or agent's file reference 00246/514W03 | | | | |
| International filing date (day/month/year) 20 October 1999 (20.10.99) | Priority date (day/month/year) 21 October 1998 (21.10.98) | | | | |
| Applicant GORDON, Roy, G. et al | | | | | |
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| X in the demand filed | with the International Preliminary | Examining Authority on: | |
| _ | 18 May 2000 (| 18.05.00) | |
| in a notice effecting | later election filed with the Intern | national Bureau on: | • |
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| made before the expiration | of 19 months from the priority d | ate or where Rule 32 applies | within the time limit under |
| Rule 32.2(b). | or ro mondio nom the phoney o | ato or, where hale oz applies, | within the time limit chaer |
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The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35

Authorized officer

C. Villet

Telephone No.: (41-22) 338.83.38

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US9924533

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PATENT COOPERATION TREATIY

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| MUDO | | | | DC. |

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

| Applicant's or agent's file reference 00246/514WO3 | FOR FURTHER ACTION | TION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416) | | | | | |
|--|---|--|--|--|--|--|--|
| International application No. | International filing date (day) | (day/month/year) Priority date (day/month/year) | | | | | |
| PCT/US99/24533 | 20 OCTOBER 1999 | | 21 OCTOBER 1998 | | | | |
| International Patent Classification (IPC) or national classification and IPC IPC(7): C23C 16/00, 16/04, 16/18; C07F 3/00 and US Cl.: 427/252; 106/1.05, 1.25; 556/40 | | | | | | | |
| Applicant THE PRESIDENT AND FELLOWS OF HARVARD COLLEGE | | | | | | | |
| This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36. This REPORT consists of a total of sheets. This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority. (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT). | | | | | | | |
| These annexes consist of a to | tal of <u>U</u> sheets. | · | | | | | |
| 3. This report contains indication | s relating to the following | items: | | | | | |
| I X Basis of the report | | | | | | | |
| II Priority | | | · | | | | |
| III Non-establishmen | t of report with regard to n | ovelty invent | ive step or industrial applicability | | | | |
| | • | overty, mvem | ive step of industrial applicationity | | | | |
| IV Lack of unity of | | - | · | | | | |
| | at under Article 35(2) with re nations supporting such state | | y, inventive step or industrial applicability; | | | | |
| VI Certain documents | cited | | | | | | |
| VII Certain defects in the | he international application | | | | | | |
| VIII Certain observation | s on the international applica | tion | | | | | |
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| Date of submission of the demand | Do | e of completion | of this report | | | | |
| Date of submission of the demand | Dat | e or completto | n of this report | | | | |
| 18 MAY 2000 | | 28 DECEMBE | R 2000 | | | | |
| Name and mailing address of the IPEA/ Commissioner of Patents and Tradem Box PCT Washington, D.C. 20231 | 1 | horized officer JEAN F VOLI | ea Jaurence To | | | | |
| Facsimile No. (703) 305-3230 | Tele | ephone No. | (703) 308-1235 | | | | |



International application No.

PCT/US99/24533

| I. Ba | sis of the r | eport | | | |
|--------------|--|---|--|--|---|
| 1 With | regard to the | elements of the internati | onal application:* | | |
| \mathbf{x} | | tional application as o | | | |
| x | the descrip | tion: | | | |
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| | pages | NONE | | | , filed with the demand |
| | pages | NONE | , filed | d with the letter of | |
| \mathbf{x} | the claims: | | | | |
| ک | pages | | | | , as originally filed |
| | pages | | , as a | | y statement) under Article 19 |
| | pages | | | | , filed with the demand |
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| [] | the drawing | 76. | | | |
| X | pages | | | , | , as originally filed |
| | pages | | | | , filed with the demand |
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| X | | e listing part of the de | = | | |
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| | the language or 55.3). | of the translation furnis | shed for the purposes | of international preliminary of | examination (under Rules 55.2 and/ |
| | | ny nucleotide and/or mination was carried o | | | nal application, the international |
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| 닏 | | ubsequently to this At | | | · |
| | furnished si | ubsequently to this At | uthority in computer | r readable form. | |
| | The stateme | nt that the subsequentl I application as filed h | y furnished written s as been furnished. | sequence listing does not go | beyond the disclosure in the |
| | The statemen | | ecorded in computer i | readable form is identical to | the writen sequence listing has |
| 4 X | The amend | ments have resulted i | n the cancellation o | f: | • |
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| | | escription, pages | | | |
| | | laims, Nos. | NONE | | |
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| in th | acement sheet. is report as | s which have been furnish | ned to the receiving Offi | mental Box (Rule 70.2(c)).** ice in response to an invitation is report since they do not co | under Article 14 are referred to ntain amendments (Rules 70.16 |
| | 70.17). replacement | sheet containing such a | mendments must be r | eferred to under item 1 and | annexed to this report |



International application No.

PCT/US99/24533

| V. | Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; |
|----|--|
| | citations and explanations supporting such statement |

| 1. | statement | | | |
|----|-------------------------------|--------|-------------------|-----|
| | Novelty (N) | Claims | 10-13 | YES |
| | | Claims | 1-9, 14-19 and 21 | NO |
| | Inventive Step (IS) | Claims | 10-13 | YES |
| | • • • | Claims | 1-9, 14-21 | NO |
| | | | | _ |
| | Industrial Applicability (IA) | Claims | 1-21 | YES |
| | | Claims | NONE | NO |

2. citations and explanations (Rule 70.7)

Claims 1-9, 14-19 and 21 lack novelty under PCT Article 33(2) as being anticipated by Gordon et al (US 5139999).

Gordon et al teaches chemical vapor deposition processes using alkaline earth metal precursors including beta diketonates (column 4, lines 4-39).

Gordon et al teaches that the precursor is in combination with an amine moiety including tetramethylethylenediamine and other amine multidentates.

Gordon et al teaches that the compounds include barium dipivaloylmethane and barium hexafluoroacetylacetonate. Gorden et al teaches that the metals can be selected from strontium and barium.

When Gordon et al teaches that the deposited material can be one or more alkaline earth metal oxides (column 5, lines 37-50) and can be deposited by various methods including spin coating then the claims are anticipated (column 5, line 27).

Claims 1, 16 and 20 lack an inventive step under PCT Article 33(3) as being obvious over Gordon et al (US 5139999).

Gordon et al teaches chemical vapor deposition processes using alkaline earth metal precursors including beta diketonates (column 4, lines 4-39).

Gorden et al teaches that the alkaline eart metal beta metal diketonate and amine are heated to a vapor and deposit a material containing alkaline metal oxides on a substrate (column 5).

Gorden et al does not specifically teach a sol-gel process. However the sol-gel process is a common process in the art for deposition of an alkaline earth metal material and Gorden et al teaches that any commonly known process can be used for the deposition. It would have been obvious to one of ordinary skill in the art to have used the process of Gorden et al and to have used the commonly known sol-gel process for the deposition.

Claim 1-3,5, and 7-8 lack novelty under PCT Article 33(2) as being anticipated by rimmer et al (US5248787).

When Trimmer et al teaches volatile barium, strontium and calcium beta diketonates with a nitrogen donor that includes tetramethyl ethylenediamine (see abstract, column 1, lines 40-68 (Continued on Supplemental Sheet.)



International application No.

PCT/US99/24533

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of: Boxes I - VIII

Sheet 10

V. 2. REASONED STATEMENTS - CITATIONS AND EXPLANATIONS (Continued): and column 2, lines 49-50)then the claims are anticipated.

Claims 1-3,5, 7-8, 14-19 lack inventive step under PCT Article 33(3) as being obvious over Trimmer et al (US5248787).

Trimmer et al teaches volatile barium, strontium and calcium beta diketonates with a nitrogen donor that includes tetramethyl ethylenediamine (see abstract, column 1, lines 40-68 and column 2, lines 49-50).

Trimmer et al teaches that the compounds are deposited on a substrate by spin coating with MO-CVD techniques (column 4, lines 1-25)to form oxides such as calcium, strontium or barium oxides.

Although the use of amine composition of beta diketonates of alkaline metals is not exemplified it is expressly suggested.

Claims 1-4 and 6 lack novelty under PCT Article 33(2) as being anticipated by Chem abs 359 (CA:118:70359).

When Chem abs 359 teaches beta diketonate compounds of barium with the triethyl amine wherein the beta diketonate is ,2,6,6-tetramethyl 3,5-heptanedione then the claims are anticipated.

Claims 10-13 the criteria set out in PCT Article 33(2)-(4), because the prior art does not teach or fairly suggest compounds and amines of the formula/composition found in instant claims 10-13.

| | NEW | CITATIONS | |
|------|-----|-----------|--|
| NONE | | | |





INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7: C23C 16/00, 16/04, 16/18, C07F 3/00

A1

(11) International Publication Number:

WO 00/23635

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(22) International Filing Date:

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60/105,158 60/126,793 21 October 1998 (21.10.98) 30 March 1999 (30.03.99)

US

(71) Applicant (for all designated States except US): THE PRESIDENT AND FELLOWS OF HARVARD COLLEGE [US/US]; 17 Quincy Street, Cambridge, MA 02138 (US).

(72) Inventors; and

(75) Inventors/Applicants (for US only): GORDON, Roy, G. [US/US]; 22 Highland Street, Cambridge, MA 02138 (US).

TEFF, Daniel [US/US]; Apartment 2007, Unit 4/28-Building 3, 7125 East Superstition Springs Boulevard, Mesa, AZ 85208 (US).

474) Agent: SCOZZAFAVA, Mary, Rose; Clark & Elbing LLP, 176 Federal Street, Boston, MA 02110-2214 (US). (81) Designated States: CA, JP, KR, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published

With international search report.

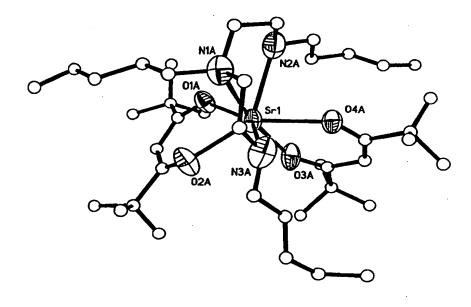
Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

(54) Title: LIQUID COMPOUNDS FOR FORMATION OF MATERIALS CONTAINING ALKALINE EARTH METALS

(57) Abstract

Q

A liquid precursor is provided for the formation of alkaline earth containing materials. The liquid precursor comprises an alkaline earth metal beta-diketonate bound to an amine. For example, a liquid compound was formed by reacting N,N',N''-trihexyldiethylenetriamine with barium 2,2,6,6-tetramethyl-3,5 heptanedionate. Films containing alkaline earth metals are deposited from vapors of the precursor liquids and, optionally, oxygen or other sources of oxygen. This process may be used to deposit barium strontium titanate films having a high dielectric constant. The liquid precursors may also be used for spray coating and sol-gel deposition The figure is an of materials. X-ray crystallographic structure of strontium bis (2,2,6,6-tetramethylheptane-3,5-dionate) with N,N', N"-triamyldiethylenetriamine.

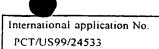


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| A. CLA IPC(7) | SSIFICATION OF SUBJECT MATTER :C23C 16/00, 16/04, 16/18; C07F 3/00 | | | | | |
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| US CL According | :427/252; 106/1.05, 1.25; 556/40 to International Patent Classification (IPC) or to both | h national classification and IPC | | | | |
| | DS SEARCHED | | | | | |
| | documentation searched (classification system follow | ed by classification symbols) | | | | |
| U.S. : | 427/252; 106/1.05, 1.25 | · | | | | |
| Documenta | tion searched other than minimum documentation to t | he extent that such documents are included | in the fields searched | | | |
| Electronic of | data base consulted during the international search (r | name of data base and, where practicable, | search terms used) | | | |
| ļ | e Extra Sheet. | · | | | | |
| C. DOC | UMENTS CONSIDERED TO BE RELEVANT | | | | | |
| Category* | Citation of document, with indication, where a | ppropriate, of the relevant passages | Relevant to claim No. | | | |
| Y | US 5,248,787 A (TIMMER et al) 28 see column 2, lines 42-57. | September 1993 (28.09.93), | 1-5, 7-8, 14-21 | | | |
| A | US 5,280,012 A (KIRLIN et al) 18 January 1994 (18.01.94), see 1-21 columns 1-6. | | | | | |
| A | US 5,225,561 A (KIRLIN et al) 06 July 1993 (06.07.93), see 1-21 columns 4-6. | | | | | |
| Y | US 5,139,999 A (GORDON et al) 18 August 1992 (18.08.92), see columns 4-5. | | | | | |
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| X Furth | er documents are listed in the continuation of Box C | See patent family annex. | | | | |
| - | cial categories of cited documents: | "T" later document published after the interdate and not in conflict with the applications and the second s | tion but cited to understand the | | | |
| to b | e of particular relevance | principle or theory underlying the invertible "X" document of particular relevance; the | | | | |
| "L" doc | ier document published on or after the international filing date ument which may throw doubts on priority claim(s) or which is d to establish the publication date of another citation or other | considered novel or cannot be considered when the document is taken alone | ed to involve an inventive step | | | |
| spec | cial reason (as specified) | "Y" document of particular relevance; the considered to involve an inventive | step when the document is | | | |
| | ument referring to an oral disclosure, use, exhibition or other means ument published prior to the international filing date but later than | combined with one or more other such being obvious to a person skilled in the | documents, such combination | | | |
| | priority date claimed | "&" document member of the same patent f | family | | | |
| | actual completion of the international search | Date of mailing of the international sear 08 MAR 2000 | ch report | | | |
| | /BER 1999 | ^ · · · · · · | | | | |
| Commission Box PCT | ailing address of the ISA/US er of Patents and Trademarks D.C. 20231 | JEAN F VOLLANO | XX9,1201 | | | |
| Facsimile No | | Telephone No. (703) 308-1235- | 7(JU ' | | | |



International application No. PCT/US99/24533

| | B. FIELDS SEARCHED Electronic data bases consulted (Name of data base and where practicable terms used): | | | | | | | |
|---|--|--|--|--|--|--|--|--|
| l | EAST, CAS ONLINE, GMELIN | | | | | | | |
| | search: dionate, barium, calcium, magnesium, strontium, gel-sol, spin-coating, spray-coating, amine, diamine, triamine, cvd, chemical vapor deposition | | | | | | | |
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INTERNATIONAL SEARCH REPORT

Inti donal Application No PCT/US 98/07829

| A. CLASSII IPC 6 | FICATION OF SUBJECT MATTER C07F19/00 C23C16/00 | | | | | |
|--|---|---|---|--|--|--|
| According to | b International Patent Classification (IPC) or to both national classif | ication and IPC | | | | |
| | SEARCHED | | | | | |
| Minimum do IPC 6 | ocumentation searched (classification system followed by classifica CO7F C23C | tion symbols) | | | | |
| Documentat | Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched | | | | | |
| Electronic d | ata base consulted during the international search (name of data t | pase and, where practical, search terms used) | | | | |
| C. DOCUM | ENTS CONSIDERED TO BE RELEVANT | | | | | |
| Category ° | Citation of document, with indication, where appropriate, of the n | elevant passages | Relevant to claim No. | | | |
| Α | CHEMICAL ABSTRACTS, vol. 103, no. 23 September 1985 Columbus, Ohio, US; abstract no. 93770, MALHOTRA, R. K. ET AL: "Synerg extraction of uranium(VI) by a noxine and beta diketones" XP002070941 see abstract & PROC INDIAN ACAD. SCI., CHI (1985), 94(3), 515-19 CODEN: PIN 0253-4134, 1985, | ism in the mixture of EM. SCI. | 1 | | | |
| X Furt | ther documents are listed in the continuation of box C. | X Patent family members are listed | in annex. | | | |
| "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filling date "L" document which may throw doubts on priority claim(s) or involve an invention which is cited to establish the publicationdate of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "at the art which is not cited to end to end the cannot be considered to exhibit the considered to exhibit the considered to exhibit the cannot be considered to exhibit the exhibit the cannot be considered to exhibit the exhibit that the exhibit the cannot be considered to exhibit the exhibit that the exhibit that the exhibit the cannot be considered to exhibit the exhibit that the exhibit th | | or priority date and not in conflict with cited to understand the principle or th invention "X" document of particular relevance; the cannot be considered novel or cannor involve an inventive step when the dc cannot be considered to involve an indocument is combined with one or ments, such combination being obvious in the art. "&" document member of the same patent | nsidered novel or cannot be considered to rentive step when the document is taken alone articular relevance; the claimed invention insidered to involve an inventive step when the combined with one or more other such docu- combination being obvious to a person skilled inber of the same patent family | | | |
| | actual completion of the international search July 1998 | Date of mailing of the International sea | агся героп | | | |
| | mailing address of the ISA European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo ni, | Authorized officer Rinkel, L | | | | |

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INTERNATIONAL SEARCH REPORT

int. .tional Application No
PCT/US 98/07829

| | PC1/US 98/U/829 |
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| | Relevant to claim No. |
| | |
| CHEMICAL ABSTRACTS, vol. 71, no. 8, 25 August 1969 Columbus, Ohio, US; abstract no. 33898, WOO, CHING-HUNG: "Extractions of several rare earths using a combination of thenoyltrifluoroacetone and acetylacetone" XP002070942 see abstract & U. S. AT. ENERGY COMM. (1968), ORO-2124-18, 94 PP. AVAIL.: DEP.;CFSTI FROM: NUCL. SCI. ABSTR. 1969, 23(7), 11605 CODEN: XAERAK, 1968, | |
| EP 0 527 661 A (THE ASSOCIATED OCTEL COMPANY LIMITED) 17 February 1993 see the whole document | 1-24 |
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| | 25 August 1969 Columbus, Ohio, US; abstract no. 33898, WOO, CHING-HUNG: "Extractions of several rare earths using a combination of thenoyltrifluoroacetone and acetylacetone" XP002070942 see abstract & U. S. AT. ENERGY COMM. (1968), ORO-2124-18, 94 PP. AVAIL.: DEP.;CFSTI FROM: NUCL. SCI. ABSTR. 1969, 23(7), 11605 CODEN: XAERAK, 1968, —— EP 0 527 661 A (THE ASSOCIATED OCTEL COMPANY LIMITED) 17 February 1993 see the whole document ———————————————————————————————————— |

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INTERNATIONAL SEARCH REPORT

information on patent family members

Int Vional Application No PCT/US 98/07829

| cited in search report date | member(s) | date |
|-----------------------------|--|--|
| EP 527661 A 17-02-1993 | AU 658922 B AU 2437692 A CA 2113715 A WO 9304072 A JP 7500318 T NO 940471 A | 04-05-1995 16-03-1993 04-03-1993 04-03-1993 12-01-1995 13-04-1994 |